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13. ABSTRACT *(Maximum 200 words)*

This report details experimental investigations undertaken to understand the interactions of aerosol particles with laser beams. Both optical and thermal interactions were examined, including elastic scattering and fluorescence, as well as a variety of nonlinear interactions. A number of new and significant experimental findings we made which are detailed in this report. These findings provide the experimental basis for potential applications in the fields of atmospheric probing, remote sensing, and photonics.

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FINAL REPORT
(Grant Number DAAL03-90-G-0176)

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I. Introduction

This is the final report for U S Army Research Office Contract Number DAAL03-90-G-0176. During the contract, a significant number of experimental and theoretical modeling investigations were carried out in my laboratory in the physics department at New Mexico State University. These investigations, which provided important insights into the basic character of aerosol processes, form the necessary first step in possible applications to atmospheric studies, aerosol science, and remote sensing. Many of these investigations were published in high-quality journals devoted to optical science. Several conference presentations were given, including invited presentations, that described the results of work performed under this contract. The Principal Investigator (PI) presented the results of this work in research seminars given at universities both in the US and abroad. The contract provided both financial and experimental facilities research support for three graduate students, and one post-doctoral student. Specifically, one graduate student received a Ph.D. and two received M.S. degrees for research supported by this contract. The post-doctoral student provided ongoing support for all experimental work including design, data acquisition and analysis, and physical interpretation.

In the remainder of this report, I will describe activities under this contract in more detail. Section II summarizes significant findings obtained during our investigations, including references to our research papers that discuss these findings in greater detail. These referenced papers are listed in a bibliography of published work under this contract, given in Section III. Section IV itemizes the conference presentations in which work performed under this contract was discussed. Finally, Section V provides information on participating scientific personnel, including the names and research topics of students obtaining advanced degrees for work performed under this contract.

II. Significant Research Findings

Significant research findings obtained under this contract may be conveniently grouped under several topical areas. These are discussed in the following paragraphs (numbered references are given in the Bibliography in Section III.). We also note potential application areas which will benefit from these studies.

II.1. Thermal Effects in Irradiated Aerosols.[1,2,16]

Irradiation of absorbing aerosols by laser beams causes a variety of thermal heating effects to arise. These effects, which depend on aerosol and laser beam properties, exhibit novel and interesting temporal characteristics. In our experimental studies, we observed several previously unreported superheating effects including the so-called Landau surface instability, which causes the aerosol surface to erupt into chaotic boiling. We also observed, for the first time, threshold behavior associated with the onset of superheating in absorbing aerosols. Findings of this nature may find applications in atmosphere probing and remote sensing.

II.2 Descartes Ring.[3,4,7,17,19]

Irradiation of transparent aerosols with a pulsed laser may generate a variety of both linear and nonlinear optical processes because of the intense pump fields inside the aerosol. In particular, near the internal focus of the pump, the intense fields may initiate electrostrictive or Kerr-effect perturbations of the refractive index which result in enhanced scattering of the pump (or of any internally generated optical signal) from the aerosol. In my laboratory, these intense focal-region field enhancements were first observed and subsequently explained; the distinctive geometry of the radiation patterns from aerosols were named the Descartes ring in spherical aerosols, and the Descartes line in microcylinders. Applications of the novel radiation effect include aerosol remote sensing and photonics.

II.3 Energy Transfer Processes.[8,10,11]

When aerosols are irradiated by electromagnetic radiation, many optical processes may be initiated. However, the optical process may not simply be the result of the interaction between an active aerosol molecule and the electromagnetic field. The process may be initiated by one of a number of so-called energy-transfer mechanisms. In aerosols, these processes are enriched by the presence of electromagnetic resonances (MDR's), which may greatly enhance a particular process. In my laboratory, we have studied energy-transfer mechanisms important in both linear processes, such as fluorescence, and nonlinear effects, such as lasing. Several new and interesting properties of these mechanisms, which were observed for the first time, result from the coupling between MDR's and the pump field.

II.4 Coherent Scattering from Seeded Microdroplets.[11]

Naturally occurring aerosols often consist of small impurities (e. g., dust particles or gas bubbles) entrained in liquid microdroplets (e. g., rain, fog, or mist). Elastic scattering from such composite system is generally noisy, with the result that the scattering signal may not be used to provide

quantitative information concerning aerosol properties. However, in my laboratory, we have investigated coherent angular elastic scattering from seeded liquid microdroplets. We have observed that the coherent spectra does not become noisy but retains its coherent profile. The effect of seeding is rather to introduce fluctuations in the amplitude of the coherent angular peaks without significantly changing their shape or separation. Temporal analysis of these fluctuations will provide detailed information concerning aerosol parameters, notably temperature. This finding will have important potential applications to remote sensing and atmospheric probing; in particular, this study will find application to the remote sensing of atmosphere temperature.

II.5 Coherent Emission from Microdroplets.[14,17,19]

The previous section deals with coherent angular elastic scattering from seeded microdroplets. However, coherent emission may also be observed from nonlinear processes in unseeded microdroplets. In particular, we have investigated coherent emission of stimulated Raman scattering (SRS) from transparent microdroplets. Although this effect was first observed by another research group, in my laboratory we measured a number of its interesting properties, and, in addition, made the first measurements of coherent microdroplet emission coupled to an external optical cavity. This effort will find potential application to a variety of remote sensing and atmospheric probing applications where the determination of the scattering phase is of interest (e. g., in Mueller matrix studies).

II.6 Enhanced Outcoupling.[17,19]

As an outgrowth of our measurements of coherent emission from laser-irradiated droplets, we have made the first measurements of controlled, enhanced emission of radiation from emitting aerosol droplets. Normal emission from irradiated aerosols occurs as a result of leakage of light from the aerosol surface. This leakage is a consequence of surface scattering (e. g., caused by thermally-induced surface roughness). However, we have found that the emission may be controlled using an optical signal, thereby effectively forming an optical switch. Furthermore, the enhanced emission exhibits a characteristic geometric signature, which may be used to distinguish it from spontaneously occurring surface emission. Controlled enhanced outcoupling will find numerous potential applications in atmospheric probing, remote sensing, and photonics.

II.7 Spectroscopy of Fractal Structures.[18]

Many atmospheric aerosols consist of fractal material entrained in liquid water droplets, such as rain, fog, or mist. For example, soot, as well as many pollutant and biological agents, are fractal in nature. The spectroscopy of fractal media is not well-understood, but recent theoretical work has indicated

that the spectra will consist of a number of high-Q resonances. If the fractal media is entrained in a liquid aerosol droplet, with its own spectrum of MDR's, spectra of the composite particle may be anticipated to exhibit many features of interest. In my laboratory, we have recently begun to make such measurements of these composite fractal media. Elastic scattering spectra taken of these media indeed exhibit interesting anfd novel features, but we have not had the opportunity to study them in detail. A variety of atmospheric probing and remote sensing applications await further understanding of this novel class of composite materials.

III. Bibliography

The following references list papers published under this contract.

1. "Evaporative Instability in Pulsed Laser-Heated Droplets," Physical Review Letters, vol. 66, p. 2988 (1991). (with J.-G. Xie, T. Ruekgauer, and R. Pinnick).

2. "Multiple Superheating Thresholds of Micrometer-Sized Droplets Irradiated by Pulsed CO_2 Lasers," Optics Letters, vol. 16, p. 1129 (1991). (with H. Latifi, J.-G. Xie, T. Ruekgauer, and R. Pinnick).

3. "Observations of Descartes Ring Stimulated Raman Scattering in Micrometer-Sized Water Droplets," Optics Letters, vol. 16, p. 1310 (1991). (with J.-G. Xie, T. Ruekgauer, J. Gu, and R. Pinnick).

4. "Physical Basis for Descartes Ring Scattering in Laser-Irradiated Microdroplets," Optics Letters, vol. 16, p. 1817 (1991). (with J.-G. Xie, T. Ruekgauer, J. Gu, R. Pinnick, and D. Pendleton).

5. "Aerosol-Induced Laser Breakdown Thresholds: Effect of Resonant Particles," Applied Optics, vol. 31, p. 311 (1992). (with R. Pinnick, A. Biswas, and J. Pendleton).

6. "Observation of Suppression of Morphology-Dependent Resonances in Singly-Levitated Micrometer-Sized Droplets," Applied Optics, vol. 31, p. 2148 (1992). (with M. Essien and J. Gillespie).

7. "Stimulated Raman Scattering and Lasing in Micron-Sized Cylindrical Liquid Jets: Time and Spectral Dependence," Journal of the Optical Society of America, vol. B9, p. 865 (1992). (with R. Pinnick, G. Fernandez, J.-G. Xie, T. Ruekgauer, and J. Gu).

8. "Energy-Transfer-Assisted Lasing from Microdroplets Seeded with Fluorescent Sol," Optics Letters, vol. 17, p. 943

(1992). (with J.-G. Xie, T. Ruekgauer, and R. Pinnick).

9. "Observations of Stimulated Raman Scattering and Laser-Induced Breakdown in Millimeter-Sized Droplets," Optics Letters, vol. 17, p. 1569 (1992). (with A. Biswas, R. Pinnick, J.-G. Xie, and T. Ruekgauer).

10. "Effects of Submicron-Sized Particles on Microdroplet Lasing," Optics Letters, vol. 18, p. 119 (1993). (with J.-G. Xie, T. Ruekgauer, J. Gu, and R. Pinnick).

11. "Suppression of Stimulated Raman Scattering from Microdroplets by Seeding with nm-Sized Latex Particles," Optics letters, vol 18, p. 340 (1993). (with J.-G. Xie, T. Ruekgauer, and R. Pinnick).

12. "Lasing Emission from an Evaporating, Layered Microdroplet," Optics Letters, vol. 18, p. 762 (1993). (with M. Essien, and J. Gillespie).

13. "Effect of Particulate Seeding on Microdroplet Angular Scattering," Optics Letters, vol. 18, p. 1293 (1993). (with J. Gu, T. Ruekgauer, and J.-G. Xie).

14. "Coherent Stimulated Raman Emission from a Microcavity-Excited Optical Resonator," Optics Letters, vol. 19, p. 58 (1994). (with J.-G. Xie, T. Ruekgauer, J. Gu, M. Young, and P. Nachman).

15. "Random Occurance of Stimulated Raman Scattering Emission from Liquid Water Microdroplets," Applied Optics, vol. 33, p. 368 (1994). (with J.-G. Xie, T. Ruekgauer, T. Gu, and R. Pinnick).

16. "Explosive Boiling of Water Droplets Irradiated with Intense CO_2 Laser Radiation: Numerical Experiments," Applied Optics, vol. 33, 1994. (with Y. Geints and A. Zemlyanov).

17. "Nonlinear Outcoupling of Stimulated Raman Scattering from Laser-Irradiated Microcylinders," Optics Letters (in press). (with J.-G. Xie, P. Nachman, and T.E. Ruekgauer)

18. "Spectroscopy of Fractal Aggregates," Physical Review (in press). (with V. Shaleav and W.-T. Kim).

19. "Lasing and Nonlinear Optical Effects in Cylinders and Scattering in Spheres," invited chapter in review volume <u>Optical Processes in Microcavities</u>, A. J. Campillo and R. K. Chang, ed., to be published by World Scientific, 1995. Manuscript in preparation.

IV. Conferences Presentations

The following conference presentations were made describing work done under this contract. They represent a mix of national and international conferences, and of invited and contributed presentations.

1. "Explosive Vaporization of Laser Irradiated Aerosols, Interational Conference on Lasers, '90, San Diego, CA, December 1990. Invited presentation.

2. "Superheating Phenomena in Absorbing Microdroplets Irradiated by Pulsed Lasers," International Conference on Nonlinear Optics and Materials, Dallas, TX, May 8-10, 1991. Invited presentation.

3. "Superheating Phenomena in Pulsed Laser Irradiated Micron-Sized Droplets," topical meeting on atmospheric, volume and surface scattering and propagation, International, Florence, Italy, August 1991. Invited presentation.

4. "Evaporative Instability in Pulsed-Laser-Irradiated Droplets," 1991 CRDEC Scientific Conference on Obscuration and Aerosol Research, Aberdeen, MD, June 24-27, 1991 (with J.-G. Xie, T. E. Ruekgauer, and R. Pinnick).

5. "Stimulated Raman Scattering in Micron-Sized Liquid Cylindrical Jets," 1991 CRDEC Scientific Conference on Obscuration and Aerosol Research, Aberdeen, MD, June 24-27, 1991. (with R. Pinnick, J.-G. Xie, T. Ruekgauer, and J. Gu).

6. "Observations of Descartes Ring Stimulated Raman Scattering in Micrometer-Sized Water Droplets,"CRDEC Scientific Conference on Obscuration and Aerosol Research, Aberdeen, MD, June 24-27, 1991. (with J.-G. Xie, T. E. Ruekgauer, J. Gu, and R. Pinnick)

7. "Descartes Ring Stimulated Raman Scattering from Liquid Microdroplets," Annual Meeting, Lasers and Electro-Optical Society, San Jose, CA, Nov. 4-7, 1991. (with J.-G. Xie, T Ruekgauer, J. Gu, and R. Pinnick). Invited presentation.

8. "Stimulated Raman Scattering and Lasing in Micron-Sized Cylindrical Liquid Jets: Time and Spectral Dependence," Annual Meeting, Lasers and Electro-Optical Society, San Jose, CA, Nov. 4-7, 1991. (with R. Pinnick, G. Fernandez, J-G. Xie, T. Ruekgauer, and J. Gu). Invited presentation.

9. "Suppression of Morphology-Dependent Resonances of a Single Levitated Laser-Irradiated Microdroplets," Scientific Conference on Obscuration and Aerosol Research, Aberdeen Proving Ground, MD, June 1992 (with M. Essien and J. B. Gillespie).

10. "Scattering in Millimeter-Sized Glycerol Droplets,"

Scientific Conference on Obscuration and Aerosol Research, Aberdeen Proving Ground, MD, June 1992 (with R. Pinnick, A. Biswas, J.-G. Xie, and T. Ruekgauer).

11. "Energy Transfer in Microdroplet Lasers Seeded with Fluorescent Sol," Scientific Conference on Obscuration and Aerosol Research, Aberdeen Proving Ground, MD, June 1992 (with J.-G. Xie, T. Ruekgauer, and R. Pinnick).

12. "Lasing from Layered Microspheres," Scientific Conference on Obscuration and Aerosol Research, Aberdeen Proving Ground, MD, June 1992. (with M. Essien).

13. "Energy Transfer Lasing Emission from Dye-Doped Microdroplets Seeded with Fluorescent Sol," Optical Society of America, Annual Meeting, Albuquerque, NM, September 1992 (with J.-G. Xie, T. Ruekgauer, and R. Pinnick).

14. "Stimulated Raman Scattering in Millimeter-Size Droplets," Optical Society of America, Annual Meeting, Albuquerque, NM, September 1992. (with A. Biswas, R. Pinnick, J.-G. Xie and T. E. Ruekgauer).

15. "Experimental and Theoretical Studies of Laser-Induced Explosions of Absorbing Microdroplets," International Conference on Lasers '92, Houston, TX, December 1992. (with A. A. Zemlyanov, Yu.E. Geints, et. al., Chistyakova, and J.-G. Xie).

16. "Suppression of Stimulated Raman Scattering from Microdroplets Seeded with nm_Sized Latex Particles," O/E Lase '93 (sponsored by the International Society for Optical Engineering, SPIE), 16-23 January, 1993, Los Angeles, CA. (with J.-G. Xie, T. Ruekgauer, and R. Pinnick).

17. "Micron-sized Droplets Irradiated with a Pulsed Carbon Dioxide Laser: Measurement of Explosion and Breakdown Thrsholds," International NATO Symposium on High-Power Microwaves, Ottawa, Canada, 2-5 May, 1994 (with A. Biswas, R. G. Pinnick, and J. D. Pendleton). Invited presentation.

18. "Optical Materials Based on Fractal Nano-Particle Composites," Materials Research Society Meeting, Boston 28 November through 2 December, 1994 (with V. Shalaev and W. T. Kim).

V. Contract Personnel Including Graduate Students

Personnel working on this contract include The Principal Investigator, Robert L. Armstrong, the Co-Principal Investigator, Paul Nachman, and a Post-Doctoral Fellow, Jing-Gang Xie. Three graduate students also performed their formal degree research under this contract. The graduate students are:

Marcelino Essien, Ph.D., 1993, Dissertation title: "Absorption and Fluorescence Effects of Morphology-Dependent Resonances of a Single Micrometer-Sized Droplet"

David DuBois, M. S., 1991, Thesis title: "Morphology-Dependent Resonances of Optically Levitated Aerosol Droplets Observed by Fluorescence and Raman Emission"

Jane Burrows, M.S., 1992, Thesis title: "Determination of the Constituents of Coal Particles using Laser-Induced Breakdown Spectroscopy"